

Docket No.: 9350-0169-0

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COMMISSIONER FOR PATENTS ALEXANDRIA, VIRGINIA 22313

RE: Application Serial No.: 09/618,044

Applicants: Werner POMPETZKI, et al.

Filing Date: July 17, 2000

For: PROCESS FOR THE HYDROGENATION OF

ACETONE
Group Art Unit: 1621
Examiner: E. Price

SIR:

Attached hereto for filing are the following papers:

REQUEST

APPEAL BRIEF W/APPENDIX (IN TRIPLICATE)

Our check in the amount of \$0.00 is attached covering any required fees. In the event any variance exists between the amount enclosed and the Patent Office charges for filing the above-noted documents, including any fees required under 37 C.F.R 1.136 for any necessary Extension of Time to make the filing of the attached documents timely, please charge or credit the difference to our Deposit Account No. 15-0030. Further, if these papers are not considered timely filed, then a petition is hereby made under 37 C.F.R. 1.136 for the necessary extension of time. A duplicate copy of this sheet is enclosed.

Respectfully submitted,

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DOCKET NO.: 9350-0169-0

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

IN RE APPLICATION OF:

Werner POMPETZKI et al : GROUP ART UNIT: 1621

SERIAL NO.: 09/618,044 : EXAMINER: E. Price

FILED: July 17, 2000

FOR: PROCESS FOR THE

HYDROGENATION OF ACETONE

REQUEST

COMMISSIONER FOR PATENTS ALEXANDRIA, VA. 22313

SIR:

Applicants respectfully request that the appeal process of the above-identified application at the Board of Appeals be reinstated.

Respectfully submitted,

OBLON, SPIVAK, McCLELLAND, MAIER & NEUSTADT, P.C.

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RE APPLICATION OF

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: GROUP ART UNIT: 1621

SERIAL NO: 09/618,044

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FOR: PROCESS FOR THE

HYDROGENATION OF ACETONE

APPEAL BRIEF

RECEIVED

TECH CENTER 1600/2900

COMMISSIONER FOR PATENTS ALEXANDRIA, VA. 22313

SIR:

The following is an appeal to the Board of Appeals concerning the decision by the Examiner to continue the rejection Claims 1-3 and 5-17 of the above-identified application as set forth in the Office Action dated October 23, 2003.

REAL PARTY OF INTEREST

Phenolchemie GmBH & Co. KG is the real party of interest of the present application.

RELATED APPEALS AND INTERFERENCES

There are no applications on appeal or in interference at the Board of Appeals and Interferences related to the present application.

STATUS OF CLAIMS

Claims 3 and 4 have been canceled. Claims 1, 2 and 5-17 remain active in the case.

STATUS OF AMENDMENTS

A response to the outstanding Office Action under the provisions of 37 CFR 1.111 has not been filed. In fact, appellants request reinstatement of the appeal to the Board of Appeals and Interferences.

SUMMARY OF THE INVENTION

The present invention is directed to a process for the hydrogenation of acetone by conducting the liquid-phase hydrogenation of acetone having a water content of less than or equal to 1.0 % by weight in at least two hydrogenation process stages, thereby preparing isopropanol product.

Support for the invention as summarized can be found on page 2, lines 24-28 and page 5, lines 10-11 of the specification.

ISSUES

Whether Claims 1-3 and 5-17 stand properly rejected based on 35 USC 103(a) as obvious over <u>Fukuhara et al</u>, U. S. Patent 5,081,321 in view of <u>Hiles et al</u>, U. S. Patent 4,626,604, further in view of <u>Sigma Catalog</u>, page 1994.

GROUPING OF CLAIMS

Claims 2 and 5-11 and 13-15 do not stand or fall together with Claim 1.

ARGUMENT

The process of the present invention is directed to the hydrogenation of acetone by conducting the liquid-phase hydrogenation of acetone having a water content of less than or equal to 1.0 % by wt in at least two hydrogenation process stages thereby preparing the desired isopropanol product in high yield and selectivity with substantially reduced byproduct production. The two essential features of the present process, that is conducted on an industrial scale, which enable the superior results of the hydrogenation of acetone are that (1) the reaction is conducted in at least two successive hydrogenation stages, and that (2) the hydrogenation of acetone is conducted under the condition of the acetone having a water content of ≤ 1 % by weight. Appellants again point out that they have presented clear evidence in the record of the case of the importance or criticality of these two process aspects of the present invention. That is, the Example of the present application on pages 8 and 9 shows the hydrogenation of acetone in two process stages using a nickel catalyst (10 % by weight Ni on a neutral (α-Al₂O₃ support). The isopropanol product is obtained in a high yield of 99.45 % while the amount of by-products is less than 100 ppm at the outlet of the second hydrogenation stage. Thus, there is a clear demonstration in the specification of the essential features of the present process which result in the production of isopropanol product in high yield and purity.

Appellants have also submitted evidence into the record in the form of the Declaration (37 CFR 1.132) executed July 4, 2001 which has been discussed on the record previously that shows the importance of the no more than 1 % by wt water content limitation in acetone starting material of the present claims and its impact on isopropanol product purity. The data in the table of the declaration, a copy of which is attached to the brief, clearly show that as the water content of acetone starting material increases, the amounts of

by-product impurities in the product isopropanol continue to increase. No such trend is shown or suggested by the cited prior art.

The Examiner cites the Fukuhara et al patent because it shows the hydrogenation of acetone to isopropanol product in a single reactor. In fact, the patent teaches a specific process of hydrogenating acetone to isopropanol in a reactor containing a fixed bed catalyst where it is stated that the direction of reaction liquid and hydrogen gas flows and the state of the catalyst are critical if the desired results of the process are to be achieved. Thus, as stated in the paragraph bridging columns 2 and 3 of the patent, it is critical to provide a cocurrent liquid/gas downflow relative to the fixed catalyst bed and to maintain the catalyst in the trickle bed state. The patent is absolutely silent as to the essential limitations of the present claims of (1) multiple hydrogenation stages for the hydrogenation of acetone to isopropanol, (2) the percentage of water contained in the acetone reactant being limited to $\leq 1 \%$ and (3) the amount of by-products of the reaction, if any. In fact, the Examiner has remarked on the record that, indeed, the Fukuhara et al patent does not teach or suggest these essential features of the present claims. Appellants therefore question, in light of these admissions, why the present claims have been rejected over Fukuhara et al at all, because it is precisely these three features that define the present invention? It should be noted that the Examiner expresses the opinion, as stated at page 5, lines 16-20 and page 6, lines 17-20, that even though Fukuhara et al is silent about the water content of acetone, which is true, nonetheless "there is no reason for one having ordinary skill in the art to believe that the <u>Fukuhara et al</u> process does not utilize acetone having a water content of less than or equal to 1.0 % considering the high conversion and yield of the isopropanol product." However, to the contrary there is a clear reason to believe that in the process described in Fukuhara et al, the content of water in the acetone reactant is of no material significance, because, as described

in the paragraph of column 2, lines 45-59 of the patent, in an embodiment of the process, the hydrogenation of acetone may be conducted in a reaction medium whose solvent component can be any one of the solvents mentioned in the paragraph, which solvents include water! If indeed water may be present in acetone in virtually any amount, there are therefore clear and compelling reasons why one of skill would conclude that to be successful in the hydrogenation of acetone, the content of water in the acetone is of no significance whatever, and that there is no particular reason why one would have to use an acetone starting material containing 1 % or less of water. Certainly in light of such a disclosure, the skilled artisan would in no way be led to employ acetone as a reactant that contains ≤ 1 % water. Appellants therefore continue to believe that the Fukuhara et al patent does not lead one of skill to the present invention.

Further, concerning the issue of the presence of water in acetone, appellants refer to their comments on page 5 of the present specification concerning the reactions shown on page 4 of the text. Here appellants state that some of the secondary reactions shown proceed with elimination of water. Accordingly, in order to suppress these reactions, i.e., increase selectivity, the addition of small amounts of water to the reaction media is conceivable. By contrast, in the present invention, in order to achieve greater selectivities of acetone to isopropanol, the amount of water must be minimized to the extent stated in the present claims.

Further, as to the matter of water in acetone, the Examiner cites the <u>Sigma Catalog</u> reference, which shows high purity acetone containing less than 0.5 % water, to make the point that one of skill in the art, knowing the availability of such high purity acetone, would have used such in an acetone hydrogenation process as now stated and claimed. To the contrary, however, all that the <u>Sigma Catalog</u> reference demonstrates is that acetone of

Reagent Grade quality, i.e., high purity acetone, is available on the market. Such high purity materials can only be obtained by special techniques that materially add to the costs of production of such materials. Indeed, the catalog markets the reagent grade acetone for \$7.00 for just 100 ml or \$17.00 for 1 liter. However, the mere knowledge that such low water content acetone exists as a specialty chemical does not lead one of skill in the art to the present invention when combined with the disclosure of Fukuhara et al, which discloses, as discussed above, that in the hydrogenation of acetone to isopropanol product in industrial scale operations where relatively large masses of acetone starting material are employed, the water content of the acetone starting material is not an issue, and indeed, the hydrogenation can be conducted with aqueous acetone! How, therefore, does the disclosure of the Sigma Catalog reference move the Fukuhara et al disclosure closer to the present invention? In other words, why would one of skill in the art purchase massive quantities of the expensive reagent grade acetone containing a very low content of water for use in the process of Fukuhara et al which does not require low water content acetone at all, especially when the reference discloses an entirely different process, and more complicated than that of the present invention, that produces isopropanol product in relatively high yield and purity? Moreover, the availability of the high purity, low water content acetone in no way suggests that such a low water content acetone would be the obvious and necessary choice by one of skill in the art for a two stage hydrogenation process (not taught by Fukuhara et al) in the large scale conversion of acetone to isopropanol.

As to the disclosure of <u>Hiles et al</u>, it is not clear to Appellants how the <u>Hiles et al</u> reference can be properly combined with the disclosure of <u>Fukuhara et al</u>, because the two patents are directed to two quite different aspects of aldehyde (ketone) hydrogenation. The aspect of improvement of the catalytic hydrogenation of aldehydes which <u>Hiles et al</u> have

achieved is to conduct catalytic hydrogenation in at least three hydrogenation zones of which a first is a first catalytic stage, the second is a hydrogenation and at least one intermediate stage including a penultimate stage, followed by a final hydrogenation step. Each stage contains a charge of hydrogenation catalyst and the stages are connected in series so that the material from one stage is fed to the next stage in series. Other factors of the multistage hydrogenation process are disclosed in column 4 of the reference. Note in this regard that the Hiles et al reference specifically comments on the inadequacies of a plural stage hydrogenation where hydrogenation is conducted in only two stages. Apparently in a two stage process the operator of a hydrogenation process would not be able to compensate adequately for a decline in catalyst activity without making certain adjustments to the process (column 5, lines 32-45). Accordingly, how would one of skill in the art reasonably combine the Hiles et al disclosure with Fukuhara et al, which specifies a single hydrogenation stage process in which an essential requirement is to maintain a catalyst bed in the trickle bed state, when the Hiles et al patent requires a plural stage hydrogenation process, preferably of three or more stages, and does not require the maintenance of a catalyst bed under trickle bed conditions? Still further, if there are in fact inadequacies of a two stage hydrogenation as stated by Hiles et al, and their solution is to provide a hydrogenation process of at least three stages in the context of the particulars of their process, how could such a disclosure lead the skilled artisan to the present invention, where although more than two hydrogenation stages can be employed in the hydrogenation process, nevertheless a two stage process is adequate with the proviso that the water content of the acetone starting material be no more than 1 %by weight? Clearly there is no teaching or suggestion in Hiles et al of this critical feature of the present process, so that, whereas Hiles et al would lead the skilled artisan to conclude that a two stage hydrogenation of acetone is inadequate, to the contrary, in the present invention,

at least two hydrogenation stages or steps are adequate as long as the water content limitation set forth in the present claims is maintained.

Appellants submit that one of skill in the art, desiring to improve the selectivity of a multistage hydrogenation process as disclosed in Hiles et al, would not use, in view of the disclosure of Fukuhara et al, trickle bed reactors under the very specific flow conditions taught in the Fukuhara et al reference. There is no motivation in either reference to modify either process with process and apparatus features of the other reference. Further, as pointed out above, no motivation is provided to adjust the water content of the acetone starting material in event acetone is selected for the hydrogenation process. Clearly, not only does Fukuhara et al teach a process quite different from that of the present invention, likewise the Hiles et al reference fails to teach or suggest the invention, and the combined patents still impart no motivation whatsoever to the skilled artisan to hydrogenate a starting acetone material containing no more than 1 % by wt water in at least two hydrogenation stages with the expectation of obtaining a highly pure isopropanol product in high yields.

As to the matter, as stated at page 7, lines 13-17 of the Office Action, that in view of the availability of low water content acetone as offered for sale by "Sigma," the Examiner is advised to consider that the offering of sale of products from a specialties chemical supply source such as Sigma is a clear indication of the sale of a material in relatively small volume amounts and not in the much larger amounts required by industry for a process such as that of the present invention where comparatively vast amounts of acetone are required for a given process. In fact, the cost of high purity acetone as offered for sale would be prohibitive of the use of the acetone product in the present process, as well as in those of Hiles et al and Fukuhara et al.

Claim 2

Given that neither cited patent of record teaches or suggests the limitations of present Claim 1, neither reference teaches the specific limitations of Claim 2 of conducting the hydrogenation of acetone in two stages, wherein the first stage is conducted at a temperature of 60 to 140° C and at a pressure of 20 to 50 bar. Withdrawal of the rejection of the claim is respectfully requested.

Claims 5 and 6

Claims 5 and 6 stand separately patentable because it is clear that because the cited patents do not teach the no more than 1 % water limitation of Claim 1, the patents could not possibly teach the narrower limitations of Claims 5 and 6. Withdrawal of the rejection of the claim is respectfully requested.

Claims 7 and 8

With respect to a preferred aspect of the present invention in which the hydrogenation catalyst employs a neutral support, it is noted that whereas <u>Fukuhara et al</u> in column 2, lines 20-32 disclose several hydrogenation catalysts, although alumina supports are mentioned, none of the disclosed supports are neutral supports. In this connection it should be observed that it is known that alumina exists in several different modifications, but only α-Al₂O₃ is a neutral support as disclosed in the present specification at page 7, line 14. As disclosed in Example 1 of the <u>Fukuhara et al</u> reference, Raney nickel is used, which is developed in an aqueous sodium hydroxide solution. Thus it cannot be concluded from <u>Fukuhara et al</u> that a neutral support material for a catalyst is contemplated.

Claim 9

The cited and applied patents do not show or suggest a two stage acetone hydrogenation process in which the hydrogenation is conducted at a temperature of 60 to 140° C and a pressure of 25 to 35 bar. Withdrawal of the rejection is respectfully requested.

Claims 10 and 11

Neither of the cited patents teaches an at least two stage acetone hydrogenation process, where, in the second stage of hydrogenation, the temperature of the second stage of hydrogenation is 60 to 140° C and a pressure of 20 to 50. Moreover, the narrower temperature range of 70 to 130° C of Claim 11 for the second stage of hydrogenation is neither taught of suggested. Withdrawal of the rejection is respectfully requested.

Claims 13 -15

Neither of the cited patents teaches anything about the amount of impurities formed during the reduction of acetone to isopropanol product. Certainly neither reference provides a teaching of a two stage hydrogenation of acetone starting material which results in an impurity content in product acetone of 300 ppm or less. Withdrawal of the rejection of the claim is respectfully requested.

In view of the comments above, appellants continue to be of the opinion that the continued rejection of the claims of the application is erroneous and should be REVERSED.

Respectfully submitted,

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FINDINGS OF FACT

- 1. The cited and applied Fukuhara et al patent discloses a process of catalytically hydrogenating acetone by feeding hydrogen gas and acetone liquid into a reactor containing a fixed catalyst bed from the top of the reactor to form a co-current gas/liquid downflow.

 There is no teaching of suggestion of a two stage hydrogenation process on a starting acetone material which contains no more than 1 % by wt water. In fact, other than the fact that the patent expressly states at column 2, lines 57-59 that water may be used as a solvent for the hydrogenation of acetone, the patent contains no discussion about the influence of water in the hydrogenation reaction disclosed. Indeed the Examiner has acknowledged that the patent makes no statement as to water content in the acetone starting material of the process of the patent.
- 2. The cited and applied Hiles et al patent discloses a continuous multi-stage hydrogenation process of reducing unsaturated organic starting materials including aldehydes, an important step of which is controlling the rate of supply of aldehyde or other unsaturated organic in the penultimate stage of hydrogenation in relation to catalyst volume so that the degree of hydrogenation in that stage is less than 100 %. Such a process does not represent the present process in which acetone starting material containing no more than 1 % water is hydrogenated in an at least two stage process resulting in isopropanol product of high purity.

CONCLUSION OF LAW

1. Claims 1, 2 and 5-17 are not rendered obvious under 35 USC 103(a) by the cited combination of Fukuhara et al, U. S. Patent 5,081,321 in view of Hiles et al, U. S. Patent 4,626,604 and further in view of the Sigma Catalog.

<u>APPENDIX</u>

CLAIMS ON APPEAL

1. A process for the hydrogenation of acetone, which comprises:

conducting the liquid-phase hydrogenation of acetone having a water content of less than or equal to 1.0 % by weight in at least two hydrogenation process stages, thereby preparing isopropanol product.

- 2. The process as claimed in Claim 1, wherein the liquid-phase hydrogenation in a first stage is conducted at a temperature of 60 to 140° C and a pressure of 20 to 50 bar.
- 5. The process as claimed in Claim 1, wherein the acetone to be hydrogenated has a water content of less than or equal to 0.5 % by weight.
- 6. The process as claimed in Claim 1, wherein the acetone to be hydrogenated has a water content of less than or equal to 0.2 % by weight.
- 7. The process as claimed in Claim 1, wherein the liquid-phase hydrogenation is conducted in the presence of a nickel containing catalyst on a neutral support.
 - 8. The process as claimed in Claim 7, wherein said neutral support is α -Al₂O₃.
- 9. The process as claimed in Claim 2, wherein the liquid-phase hydrogenation is conducted at a temperature of 70 to 130° C, and a pressure of 25 to 35 bar.
- 10. The process as claimed in Claim 1, wherein the liquid-phase hydrogenation in a second stage is conducted at a temperature of 60 to 140° C and a pressure of 20 to 50 bar.
- 11. The process as claimed in Claim 10, wherein the liquid-phase hydrogenation is conducted at a temperature of 70 to 130° C.
- 12. The process as claimed in Claim 1, wherein the hydrogenation is conducted at a molar ratio of hydrogen to acetone ranging from 1.5:1 to 1:1.

- 13. The process as claimed in Claim 1, wherein the total concentration of by-products formed in said liquid-phase hydrogenation reaction is less than 300 ppm.
- 14. The process as claimed in Claim 13, wherein said total amount of by-products is less than 200 ppm.
- 15. The process as claimed in Claim 14, wherein said total amount of by-products is less than 100 ppm.
- 16. The process as claimed in Claim 1, wherein the liquid-phase hydrogenation reaction is conducted in the presence of a hydrogenation catalyst of copper, chromium, ruthenium or nickel on a Al₂O₃, TiO₂ or ZrO₂ support.
- 17. The process as claimed in Claim 1, wherein the liquid-phase hydrogenation reaction is conducted in the presence of a hydrogenation catalyst of a catalytically active metal on a neutral support selected from the group consisting of α -Al₂O₃, TiO₂, ZrO₂ or mullite.